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THE

AMERICAN NATURALIST

VOL. XXIX. April, 1895. 340

ON THE PRESENCE OF FLUORINE AS A TEST FOR THE FOSSILIZATION OF ANIMAL BONES.

By Dr. Thomas Wilson.

The discovery of the existence of fluorine in animal bones was made by Morichini in the early years of the nineteenth century, and was soon thereafter confirmed by the leading chemists, although there were considerable discussions and some disputes in regard to its presence. Berzelius in his *Traitè de chimie*, Vol. VII, page 473, 1833, reports having certainly found it in animal bones, but says it was less than two per cent. About the same time, Gay Lussac, in his lectures before the Faculty of Science, says that the fluoride of calcium exists in the proportion of about one one-hundredth in the composition of bone and the enamel of teeth. On the contrary, in 1843, Girardin and Priesser, in their *Memoire sur les os anciens et fossiles*, in the *Annales de physique et de chimie*, 1843, Vol. III, page 370, declare that they have not been able to find the least trace of fluoride of calcium in ancient human bones, and that the existence of this salt in recent animal bones is more than doubtful. But this difference was all put to rest and beyond doubt by the investigations of Mons. Fremy in his *Recherches chimiques sur les os, Annales de physique et de chimie*, 1855, Vol. I, page 47. But it was, as they say, in a quantity so small that the most delicate mechanism and operation were required to find it, and emphasizing this on page 70, he declines to give any analytic method by which he can determine the exact quantity of this salt in the bones tested. The results of analyses of Von Bibra were to the same effect. Heintz believes that he had found in the ash of the human femur from three to five per cent of the fluoride of calcium; while Zaleski made it less than one per cent.

The discoveries of prehistoric man in later times has rendered this investigation of greater interest than ever before. No prehistoric anthropologist of serious application, but has greatly desired some test which might even, in a measure, determine the relative, if not the actual, antiquity of bones found in prehistoric graves. To these persons it will be welcome news that the discoveries of the existence of fluorine in different proportions in recent bones and those of antiquity will furnish a test of their respective ages, however slight and uncertain it may be. It may be only partial, cannot be depended upon in all cases, but will be, nevertheless, a source of gratification if it be a step towards the solution of a controverted point. It may be freely conceded that animal bones deposited in one locality may possess a greater quantity of flourine than they would have if deposited in another. The condition of their surroundings, the presence and association of different salts may produce a decided effect upon the results. But there may be cases in which these differences do not exist, and hence, this test may be invoked with considerable benefit. A deposit of human and animal bones may be mingled together, the fossilization of the latter may be determined from other knowledge of their antiquity, while the deposit, being in the same locality, subjected to the same conditions, having the presence of the same chemical constituents, the difference in fluorine of the various bones may afford in some degree, possibly slight, a test as to the antiquity of human bones. Such a test and comparison was sought to be made by the writer in the case of the deposit of animal bones found at Natchez by Dr. Dickeson. Along with them was a fragment of a human pelvis. While they were not actually touching each other, they were practically so—they were in the same position, same deposit, and, so far as is possible to determine, subject to the same chemical influences: therefore, a comparison between the two ought to give an idea of their comparative ages.

These bones are shown in one of the alcoves of the Museum of the Academy of Natural Sciences, Philadelphia. They are fossil bones of extinct animals of the plistocene period. In color, texture and general outward appearance they have a remarkable similarity as though they belonged together. They are well preserved, firm in texture, and of a dark chocolate-brown color which has been attributed to ferruginous infiltration. They consist of a nearly entire skull of *Megalonyx jeffersonii*, teeth of the *Megalonyx dissimilis* and the *Ereptodon priscus*, bones of the *Mylodon harlanii*, bones and teeth of the *Mastodon americanus*, and teeth of *Equus major* and *Bison latifrons*. Along with them is the os innominatum of a human subject. The question affecting the antiquity of man is whether these subjects, the bones of which were found together, were, when alive, contemporaneous, and whether the evidence of age in one is evidence of age in the other. They were all presented to the Academy by Dr. Dickeson at the meeting in October, 1846; description thereof is to be found in the Proceedings of the Society for that year, vol. iii, p. 106. Dr. Dickeson reported at that time that they were discovered by him in a single deposit at the foot of the bluff in the vicinity of Natchez, Mississippi. He says, " The stratum that contained these organic remains is a tenacious blue clay that underlies the diluvial drift east of Natchez, and which diluvial deposit abounds in bones and teeth of the *Mastodon giganteum;* that they could not have drifted into the position in which they were found is manifest from several facts, first that the plateau of blue clay is not appreciably acted on by those causes that produce ravines in the superincumbent diluvium; second, that the human bone was found at least two feet below the three associated skeletons of the *Megalonyx*, all of which, judging from the position or proximity of their several parts, had been quietly deposited in this locality, independent of any active current or other displacing powers; and lastly, because there is no mixture of diluvial drift with the blue clay, which latter retains its homogenous character equally in the higher parts which furnished the extinct quadrupeds and in its lower part which contained the remains of man." These specimens thus found associated were made the subject of investigation by Sir Charles Lyell, and afterwards by Dr. Joseph Leidy, the latter having published a memoir with illustrations of the human bone in the Transactions of the Wagner Free Institute of Science, vol. ii, p. 9. He says, "It differs in no respects from an ordinary average specimen of the corresponding recent bone of man."

Dr. Leidy says Lyell expressed the opinion that, although the human bones may have been contemporaneous with those of the extinct animals with which it has been found, he thought it more probable that it had fallen from one of the Indian graves and had become mingled with the older fossils which were dislodged from the deeper part of the cliff, and Dr. Leidy adds: "In the wear of the cliff the upper portion, with the Indian graves and human bones, would be likely to fall first, and the deeper portions with the older fossils, subsequently on the latter."

Although Dr. Leidy testifies to the general similarity of appearance of the human with the other bones, it does not seem to have occurred to him to have them analyzed and compared. Remembering the story told by the analysis and consequent comparison of the Calaveras skull with that of the rhinoceros teeth found in a formation corresponding in age, though in a different locality; and of the fact apparent therefrom that the Calaveras skull was in an equally advanced stage of fossilization as the rhinoceros teeth, I deemed it wise to make an examination and test by analysis. To this end I applied to Prof. Angelo Heilprin, and through him to the authorities of the Philadelphia Academy of Natural Sciences, so a few months since specimens certified by Prof. Heilprin have been taken, one from the bone of the man and the other from one of the bones of the mylodon, choosing those which, for size, texture and general appearance, bore the greatest likeness one to the other. These were submitted by me to Mr. Hildebrand, a Chemist of the United States Geological Survey, on duty at the National Museum, who furnishes the following result of his analysis.

TWO FOSSIL BONES.

		Man. Per cent.	*Mylodon.* Per cent.
Loss at 100°C.		4.55	6.77
Loss on ignition		16.54	21.18
Silica	(SiO_2)	22.59	3.71
Phosphoric acid	(P_2O_5)	17.39	23.24
Alumina	(Al_2O_3)	3.21	4.02
Iron protoxide	(FeO)	5.65	4.44
Manganese protoxide	(MnO)	1.65	3.40
Lime	(CaO)	25.88	30.48
Magnesia	(MgO)	0.95	0.78
		98.41	97.02

"Alkalies, carbonic acid and fluorine were not looked for, owing to the small amount of available material, hence the low summation."

The failure to test for fluorine is much to be regretted, and was the cause of a second application for a greater quantity with which to make another analysis. It will be reported further on in this paper.

The importance of this analysis will be apparent at a glance. The human bone is in a higher state of fossilization than is that of the Mylodon. It has less lime and more silica. In their other chemical constituents they are without any great difference. Of lime the bone of the Mylodon has 30.48%, while that of man has but 25.88%. Of silica the Mylodon has 3.71%, while man has 22.59%. I am well aware of the ordinary uncertainty of this test when applied to specimens from different localities and subjected to different conditions; but in the present case no such differences exist. The bones were all encased in the same stratum of blue clay, and were subjected practically to the same conditions and surroundings. As one swallow does not make a summer so the discovery of one specimen does not prove the antiquity of man; but it is to be remarked that upon each discovery and in almost every investigation the evidence found points towards higher antiquity of man and tends to show the occupation of the earth by prehistoric man to be more and more extensive. This discovery is simply a fact to be put down to the credit of the high antiquity of man. We should proceed in the same direction to discover other evidences, to investigate the value of those already found; and as they accumulate, each one, or all together, should be given their fair value, in the endeavor to arrive at a truthful conclusion, independent of *a priori* theory or preconceived idea.

The possibility of the determination of the relative antiquity of human bones found in prehistoric graves has set the chemists and prehistoric anthropolgists to an investigation of the fluorine test. Mons. Zaborowski presented a paper before the Academy of Sciences at Paris on the 1st of May, 1893, upon the differences in the chemical composition between two skeletons alleged to be prehistoric, those of Thiail and of Villejuif. These analyses were made by Mons. Adolphe Carnot, Engineer-in-Chief of Mines, and a professor of the Superior School of Mines, Paris. The result of these analyses was to show that the skeleton of Villejuif was much the oldest of the two and that it accorded well with other ancient bones which, from their surroundings and associations, were definitely determined. In the Bulletins de la Societe d'Anthropologie of Paris, Vol. IV, No. 6, the Seance of 18th of May, page 309, Mons. Emile Rivière gave the results of his investigations, comparisons and analyzations of alleged prehistoric bones belonging to the deposit of Billancourt. The excavations of this deposit have been pursued by Mons. Rivière during the seven years from 1875 to 1882, and the extinct fauna which he had obtained therefrom consisted of *Elephas primigenius*, *Rhinoceros tichorinus*, *Cervus megaceros*, *Tarandus rangifer*, *Bos primigenius*, etc. There were also found in one of the river drifts in the immediate neighborhood of Billancourt, the remains or fragments of two human skeletons. These human bones were transmitted to the museum with the suggestion that they belong to the same epoch and, consequently, were of the same antiquity as the deposit of Billancourt, although MM. Rivière and Gaudry, judging from the aspect, texture, density and, in fact, in all their physical characters, were of a contrary opinion. This last proposition was combatted by several members of the Congress, and it was insisted that the human bones were contemporaneous with the ancient animal bones. These bones were afterwards, and for the purpose of settling or aiding at least in settling the disputed question, submitted to Mons. Carnot, who reported in his analysis that fluorine in the human tibia was but $\frac{17}{100}$ of one per cent, while in the animal bones it was found in quantities of one per cent and $\frac{43}{100}$. Mons. Carnot reports that, taking the average of the specimens, the presence of fluorine in the animal was from seven to nine times greater than that in the human skeleton, and that the human skeleton contains nothing or but slightly more than was to be found in modern bones; and his conclusion was that the human bones were much more recent. Mons. Carnot has given the results of his investigation, the manner of the conduct of his experiments, his method of analysis, and his opinion of the whole matter in a recent paper in the *Annales des Mines*, 2nd Bulletin of 1890, page 155.

The remainder of this paper is, for the most part, a translation of that of Mons. Carnot which, made by the present writer, has very kindly been revised for correction of chemical terms and formulæ by Prof. R. L. Packard.

Process of Analysis pursued and recommended by M. Carnot.—Bones are composed partly of mineral and partly of organic matter. We are to deal only with the mineral, but must first determine the amount or proportion of the organic matter, and to this end the specimen to be operated upon is calcined at a moderate temperature, in such way as to preserve only the ashes or burnt bones. The caustic lime produced by calcination is converted into carbonate of lime by the addition of carbonate of ammonia, evaporation and calcination near 200 degrees before weighing the ashes. By taking the weight of fragments of bone well washed and dried, then the weight of the ashes, the difference may be considered as representing the the weight of organic matter.

The ash of modern bones is formed of the following elements: tribasic phosphate of lime in the largest proportion, with a small quantity of phosphate of magnesia, very little fluoride and chloride of calcium, carbonate of lime, very little peroxide of iron and of aluminum and, finally, an extremely small proportion of alkaline salts, as chlorides, sulphates and, possibly phosphates.

In the fossil bones we will see that the proportions of phosphate and of carbonate of lime vary, that those of magnesia and of chlorine change very slightly, while the fluoride of calcium is in a greater and more important quantity. There is often much more oxide of iron and sometimes phosphate of iron, sometimes crystallized silica, frequently a little sand or clay, and finally, in certain cases, sulphate of lime and pyrites.

Complete Analysis.—This operation, performed upon modern bones, comprises the following:

1.—Determination of fluorine.
2.—Determination of chlorine.
3.—Determination of carbonic acid.
4.—Determination of phosphoric acid and the bases (lime, magnesia, alumina, and oxide of iron).

1. *Fluorine.*—The determination of fluorine is performed in the method described in detail in the *Annales des Mines*, 1st number; 1893, page 130; the general features of which, however, are as follows: The decomposition of the fluoride in presence of a great excess of silica by concentrated sulphuric acid, in an apparatus entirely dry, with a slow current of dry gas running through it; the transformation of gaseous fluoride of silicon into fluosilicate of potassium, which is collected, washed, dried and weighed. If the chlorine is in a quantity not to be neglected, it is absorbed by passing the current of gas through pumice impregnated with sulphate of copper and heated until the dehydration is complete. The same reagent

will arrest sulphuretted hydrogen when the calcined matter contains a sulphide which is decomposed by sulphuric acid. If there be organic matter which can evolve sulphurous acid, the latter can be retained by the use of caustic lime freshly calcined. In the case of substances containing iodine, it is moved by means of the copper turnings placed before the sulphate of copper. In determining fluorine, 5 grams of the ash of modern bones to 2 grams of fossil bones are employed in such way as to always have a convenient quantity of fluosilicate of potassium to determine on the balance.

2. *Chlorine.*—For the determination of chlorine a special operation is performed, treating one or two grams of ash by cold nitric acid slightly diluted. When the solution is complete, we add water and nitrate of silver. The weight of chloride of silver dried, will give the proportion of chlorine.

3. *Carbonic Acid.*—The proportion of carbonic acid enabling us to calculate the carbonate of lime is obtained by attacking from two to four grams of ash by nitric acid in a small flask exactly weighed and provided with a tube containing pumice saturated with sulphuric acid (*paunce*) to retain humidity. At the end of the operation, the carbonic acid is replaced by dry air by aspiration, and the apparatus is weighed. The loss of weight represents the carbonic acid disengaged.

4. *Phosphoric Acid, Lime, Magnesia, Alumina, and Oxide of Iron.*—Two grams of ash are used in this operation. Dissolve in hydrochloric acid and heat in a capsule of platinum a sufficient length of time to drive out the fluorine. The solution diluted with water, is then filtered, afterwards saturated by ammonia until the commencement of precipitation of phosphate of lime, then redissolved by acetic acid added little by little in slight excess in such a manner as not to produce a noticeable heating of the liquid.

Phosphate of iron is precipitated in this way alone or with phosphate of aluminum. If this precipitate is almost insignificant, we take the weight after washing and ignition and according as (1) the absence or (2) the presence of aluminum is observed, we calculate by the following formula the weight of the different elements (without separating the iron and aluminum):

$$\text{(1.)}\ P_2O_5Fe_2O_3=p,\quad P_2O_5=0{,}47p,\quad Fe_2O_3=0{,}53p.$$
$$\text{(2.)}\ P_2O_5\binom{Fe_2O_3}{Al_2O_3}=\acute{p},\ P_2O_5=0{,}\ 50\acute{p},\binom{Fe_2O_3}{Al_2O_3}=0{,}50\acute{p}.$$

If the precipitation is somewhat voluminous, the elements are determined by separating the phosphoric acid by molybdate of ammonia. The acetic liquid, from which phosphate of iron and aluminum has been removed, is diluted to 200 cubic centimetres. Exactly the half is then taken to determine the lime, the magnesia and the phosphoric acid; the other half is put in reserve to verify the result in case of need. The addition of oxalate of ammonia, neutral and pure, will determine the precipitation of lime, which is separated as oxalate and is determined as caustic lime, carbonate or sulphate of lime. The filtered liquid, supersaturated with ammonia, is left in repose during 24 hours and gives a precipitate of ammoniomagnesium phosphate, of which the weight, after ignition, enables us to calculate the magnesia and a part of the phosphoric acid.

$$P_2O_52MgO=p_1,\quad 2MgO=0{,}36p_1,\quad P_2O_5=0{,}64p_1$$

The rest of the phosphoric acid is precipitated in the final solution by the magnesia mixture, and is determined as P_2O_52MgO.

Rapid Methods.—In the large series of modern and fossil bones which have been examined, it would have taken a long time and been, perhaps, without benefit to make the complete analysis. In most cases it is sufficient to make, along with the qualitative analysis, the exact determination of fluorine as fluosilicate of potassium and the rapid determination of phosphoric acid by means of a standard solution of uranium. This operation is executed in the following manner: 2 grams of the ash are dissolved in hydrochloric acid, and the liquid is neutralized and the phosphate of iron or iron and aluminum separated as before described, of which we take 47 or 50 per cent. to represent the weight of the phosphoric acid. Almost all the phosphoric acid is found in the acetic solution with which we proceed to a volumetric test. It is unadvisable to pour into the acetic solution the standard solution of uranium, because the latter will cause some phosphate of lime to precipitate with the phosphate of uranium and we should, in consequence, find too small a proportion by measuring the volume of uranium solution employed.

We arrive at a more exact result by making the determination by the indirect method: We dilute the liquid to 200 cubic centimetres, then pour it, with the aid of a graduated burette, into a measured volume of a standard solution of nitrate of uranium, to which is added a little acetate of soda slightly acidified by acetic acid and the solution is heated to near 80 degrees. We stop when one drop of the liquid, placed on a porcelain saucer, in contact with a little ferrocyanide of potassium in powder, will cease to give a brown color. In order to standardize the solution of uranium, we operate in the same manner, starting with an exactly known weight of phosphate of lime. By taking phosphate of lime exactly analyzed, we succeed better than by taking pure phosphate of soda, because we standardize and make the determination in conditions which are identical. We add pure water to such extent that 40 cubic centimetres correspond exactly to 0.200gr. of phosphoric acid (or 1 cubic centimetre to 0,005gr. of P_2O_5). The operation should be performed a second time for verification. In the assay of a phosphate, if one operates on 2 grams and makes the acetic solution up to 200 cubic centimetres, we can calculate the proportion of phosphoric acid from the volume V of the liquid which will have saturated the 40 cubic centimetres of the solution of uranium. The volume contained 0.200gr. of phosphoric acid. As a consequence, in 1 gram of phosphate, or 100 cubic centimetres of the liquid, there would be 0gr, $200 \times \frac{100}{V}$ (V being expressed in cubic centimetres).

The following table gives the results of analyses for a certain number of bones of varied origin.

These analyses confirm at nearly all points the conclusions that M. Fremy has drawn from his numerous analyses. We remark with him as to the constancy in the proportions of phosphate and carbonate of lime in genuine bones, whether we compare the different parts of the same bone, the different bones of the same individual, or the bones of animals of different species. The same observation can be made for the proportion of fluoride of calcium which appears always to fall within the limit from $\frac{1}{3}$ to $\frac{1}{2}$ per cent. Chloride of calcium is almost always in less quantity than fluoride; it is the same with the oxide of iron except in the shell of the turtle trionyx (IX). It is necessary, nevertheless, to notice some exception in this constancy of proportion in bony substances of special nature, like the teeth or tusk of the elephant and the shell of the turtle. The phosphate of magnesia, almost always less than 2 per cent, is raised to near 4 per cent in the tooth of the elephant (VI) and passes 15 per cent in the tusk of the elephant (VII) of which it contributes, possibly, to augment the compactness and resistance. At the same time the carbonate of lime which often forms more than 9 per cent of the substance of bone, drops to 2 per cent in the ivory. The carbonate of lime is sensibly in superior proportion in the average of bones of herbivorous animals, and above all, in those that live in water, the manatee and the turtle (IV, IX, X).

	Human—Body of Femur.	Human—Head of Femur.	Ox, Tibia.	Manatee rib—Modern.	Elephant Femur—Modern (Siam).	Elephant tooth, Dentine.	Elephant tusk, Ivory.	Bones of Grand Serpent.	Trionyx Shell.	Testudo radiata.	Crocodile.	Varan a deux Bandes (Monitor).
	I.	II.	III.	IV.	V.	VI.	VII.	VIII.	IX.	X.	XI.	XII.
$P_2O^5 3CaO$	87,45	87,87	85,72	81,82	90,03	86,87	82,08	89,80	80,37	81,87	87,67	87,32
$P_2O^5 3MgO$	1,57	1,75	1,53	2,62	1,96	3,82	15,72	1,91	1,46	1,49	1,05	1,69
$CaFl_2$	0,35	0,37	0,45	0,63	0,47	0,43	0,20	0,43	0,27	0,35	0,33	0,57
$CaCl_2$	0,23	0,30	0,30	0,36	0,20	0,20	traces	0,20	0,10	0,16	0,34	0,24
$CaOCO^2$	10,18	9,23	11,96	14,25	7,27	7,27	2,04	7,41	16,58	15,80	10,69	9,88
Fe^2O^3	0,10	0,13	0,13	0,15	0,15	0,15	0,08	0,15	1,09	0,13	0,17	0,12
	99.98	99,65	100,09	99,83	100,08	100,11	100,12	99,90	99,87	99,80	100,25	99,82

The principal object of this paper has been to compare modern bones with fossil bones of similar species in order to discover the relative amount of fluorine and phosphoric acid, and to that end numerous analyses have been made, the results of which can better be given in the form of a table.

The first column of figures gives the proportion of phosphoric acid, the second the proportion of fluorine, the third gives the calculated *ratio* of this proportion of fluorine to that of an apatite having an equal amount of phosphoric acid.

Bones of Modern Animals.	Phosphoric Acid.	Fluorine.	Proportion calculated for apatite.
Human Femur—Body	40,91	0,17	0,05
Human Femur—Head	41,20	0,18	0,05
Human rib	36,80	0,22	0,06
Bone of Ox	40,10	0,25	0,07
Bone of Calf	40,32	0,23	0,06
Bone of Elephant	41,24	0,23	0,06
Tooth of Elephant—Dentine	41,77	0,21	0,05
Modern Ivory	46,12	0,10	0,02
Bone of Manatee	38,93	0,31	0,09
Bone of Dugong	38,03	0,27	0,08
Bone of Rytina gigas	39,52	0,24	0,07
Bone of White Swan	38,20	0,19	0,05
Shell of the Trionyx	37,54	0,13	0,04
Bone of Testudo radiata	38,30	0,17	0,05
Bone of large Serpent	42,16	0,21	0,05
Bone of Crocodile	40,88	0,16	0,04
Bone of Varanus	40,91	0,28	0,07
Bone and Spine of Turbot	40,96	0,11	0,05
Bone and Spine of Codfish	41,27	0,24	0,06
Bone and Spine of Pike	38,26	0,16	0,05

It can be seen from this table that the amount or quantity of Fluorine in these modern bones is always between 0,16 and 0,31 p. 100, while the amount of phosphoric acid in the same is between the limits of 37 and 42 p. 100 (the ivory being an exception). The relation between the amount of fluorine with that of phosphoric acid in the normal apatite is found to be between 0,05 and 0,07 per cent, the exact average for the 20 analyses being 0,058.

Fossil Bones.—The following tables give the result of corresponding tests upon fossil bones, for comparison and to observe the modification produced by fossilization:

Complete Analysis.	1· Bone of Manatee, found in the peat deposits of Scania, Sweden. Plistocene Geol. Epoch.	2. Bone of Ox, from a Plistocene deposit in Cindre, Department of Allier, France.	3. Bone of Manatee from a Tertiary deposit, Tongrien stage, at Etrechy, Seine et Oise	4. A Vertebra of the Ichthyosaurus, from the Kimmeridgian clays of the Jurassic period; Havre, Seine Inferieure.	5. A silicified bone of a reptile from Bayreuth,* belonging to the Triassic period.	6. Debris of Fish from a breccia in the inferior silurian of Canon City, Rocky Mts., U. S. A.
Phosphate of Lime	64,63	79,05	68,53	12,90	....	67,70
Phosphate of Magnesia	1,97	0,65	2,27	....	....	....
Phosphate of Iron $(P_2O^5F^2eO^3)$	9,63	1,34	8,68	1,23	0,62	....
Peroxide of Iron	....	....	....	....	....	7,10
Fluoride of Calcium	0,88	1,70	3,82	3,42	traces	5,31
Chloride of Calcium	0,44	0,48	0,30	....	....	....
Carbonate of Lime	21,20	15,98	15,68	51,56	....	15,01
Silica	0,75	0,10	0,35	....	....	....
Sulphide of Iron (FeS^2)	....	....	....	24,92	....	....
Sulphate of Lime	....	....	....	4,23	....	....
Silica and Clay	....	....	....	1,57	....	....
Quartz	....	....	....	....	98,45	....
Organic matter	....	....	....	....	....	5,67
Loss on ignition	....	....	....	....	0,88	....
Total	99,50	99,30	99,63	99,83	99,95	100,79

*Note.—There remained of this only a thin crust of phosphate of iron, the central part was formed of crystals of quartz enveloped in a gray-blue quartzose mass, traversed by gray veins that served to bind together the phosphated envelope.

It is to be remarked that the first four analyses report only the mineral portion of the substance, the organic matters having first been abstracted; while in the last two, the organic matter has been reported. The relative proportions between the fluorine and phosphoric acid will be set forth further on in these tests or assays and are classed according to their geologic deposits.

It is deemed proper to cite other tables made by other chemists and reported from standard works. This for the purpose of comparison:

Fossil Bones after Girardin and Preisser, reported in the *Annales de Physique et de Chimie*, 1843, Vol. 3, p. 370.

	Hygroscopic Water.	Organic matter.	Phosphate of Lime.	Phosphate of Magnesia.	Phosphate of Iron.	Carbonate of lime.	Fluoride of Calcium.	Silica.	Alumina.	
1. Metacarpal of Fossil Bear, Cavern of Mialet, Gard	1.30	7.17	75.45	2.81	………	12.18	1.09	………	………	100
2. Tusk of Fossil Elephant, Alluvial deposit at Saint Pierre sur Dives. (Calvados.)	………	traces	75.91	3.15	………	18.40	2.64	………	………	100
3. Vertebra of the *Plesiosaurus dolichodeirus* from the Oxfordian clay at Dives, Jurassic	2.20	4.80	54.20	4.61	6 40	10.17	2.11	9.21	6.30	100
4. Large bone of the *Poekilopleuron bucklandii*, quarries of Maladrerie, calcaire of Caen, Jurassic	………	1.30	71.12	………	0.12	25.31	0.86	1.29	………	100
5. *Ichthyosaurus*—Rib-bone, from the Dives clay	………	1.34	46.00	1.00	16.11	31.09	1.02	3.21	………	100
6. *Ichthyosaurus*—Rib-bone, from the chalk chlorite	………	8.19	76.00	1.08	0.70	10.00	0.12	3.01	………	100
7. *Ichthyosaurus*, skull, from the Jurassic	0.60	7.07	70.11	1.45	………	17.12	1.65	2.00	………	100
8. Bone of Manatee, from tertiary deposits near Valogne (Manche)	………	………	76.40	………	5.71	0.97	9.12	7.80	………	100
9. *Otaria jubata* (?) Straits of Magellan, S A.	………	1.17	61.19	0.95	………	14.48	22.21		………	100

It is to be noted that the last number given for the fluoride of calcium shows that the exactitude of these analyses are not always to be depended upon.

Fossil Bones after Mons. Fremy. *Annales de Physique et de Chimie,* 1858, Vol. I, p. 88.

	Ash.	Phosphate of Lime.	Phosphate of Magnesia.	Carbonate of Lime.	Silicious matters, and Fluoride of Calcium.	Organic matter.
1. A fossil Ox from Caverns of d'Oreston: the metatarsus with exterior part having the aspect of wood	80.4	71.1	1.5	11.8	....	10.3
2. The same—interior having become friable	80.6	71.5	1.7	11.3	....	11.0
3. The same—spongy part	84.2	6.33	1.2	5.2	17.2	8.0
4. Vertebræ of fossil Rhinoceros from Sansan, (Gers.).	83 4	5.90	....	41.3	2.6	traces
5. The same—ribs	83.1	6.68	....	27.5	1.4	traces
6. Long Bone of fossil Hyæna from Cavern of Kirkdale	75.5	72.0	1.3	4.7	....	20.0
7. Dorsal Vertebra of fossil Rhinoceros	69.5	25.7	0.4	57.5	8.5	
8. Humerus of same	73.0	32.4	0.4	64.0	6.2	....
9. Teeth of same	90.4	65.2	0.7	13.8	14.5	....
10. Tusk of the fossil Mastodon	90.4	56.5	0.7	13.1	24.3	....
11. Bone of fossil Bear—solid part	83.9	59.7	0.4	23.6	9.8	....
12. The same—spongy part	76.7	23.1	1.2	67.5	14.0	....
13. Fossil Armadillo, scales	80.7	55.0	0.4	23.8	12.4	....
14. *Anoplotherium*, fossil—vertebra of the tail	84.0	53.1	0.4	20.4	19.4	....
15. Vertebra of the fossil Turtle	87.0	61.1	0.7	10.6	18.6	....

Observations.—The present composition of these fossil bones results both from their initial composition at the epoch when they belonged to the living animals, and the alterations incident to the diverse influences of atmosphere, soil and water during a long series of centuries. Comparing the results of these analyses of different modern animal bones, results with but feeble divergencies, we are authorized to believe that it was the same in past time and that the initial composition of the bone was practically uniform. The influences which have operated upon the fossil bones since they were buried in sedimentary deposits have produced modifications which are more or less important and quite varied. The proportion of organic matter has always been much diminished; from 35 to 40, which seemed to be the original proportion, it has descended in the process of fossilization from 6 to 12 per cent. These differences testify at the same time to the permeability of the earth by air and water, of infiltration and to the degree of compactness or porosity of the different bones or to the different parts of the same bone. The modification in the organic matter of the bones has not been examined, but the reader is recommended to an interesting study upon that subject by Mons. Scheurer-Kestner in the *Bulletin de la Societe chimique*, 1870, Vol. I, page 199, and Vol. II, page 11.

As for the mineral substances of these bones, the difference is sometimes small, but other times large from the initial composition. The proportion of carbonate of lime is generally increased, although in a measure extremely variable. The amount of phosphate of lime has frequently diminished, whether by reason of solution and infiltration of water or by the formation of phosphate of iron. The magnesia is found in proportion nearly identical to those which exist in the bones of living animals. The bones have always retained a notable quantity of iron, whether in the state of hydrate of peroxide which colors them a reddish-brown, or in the state of phosphate of peroxide, rarely in the state of ferrous phosphate. The reducing actions have sometimes determined the deposit of sulphide of iron under the form of pyrites inattackable by hydrochloric acid. This has taken place notably in the vertebra of the *ichthyosaurus* from the Kimmeridgian clays of Havre. There is sometimes found in these bones, a certain quantity of sulphate of lime. The calcination with organic matter could give rise to the sulphur. The bones contain frequently a small quantity of clay or of silica in the form of quartz. They are also, though rarely, almost entirely transformed into crystalline or crystallized quartz; such is the case with the specimen from the trias at Bayreuth.

The chlorine is almost always in slight proportion in the fossil bones as well as in the modern, especially if one has taken care to eliminate by washing the soluble chlorides, and to leave only the chloride of calcium combined with the phosphate of lime. Fluorine is found always in a greater and more important quantity corresponding to several hundredths of fluoride of calcium. This salt forms with the phosphate of lime a nearly insoluble compound comparable to apatite; it appears to augment, in a certain measure, with the antiquity of the bones, so that one is able, up to a certain point, to find in the relative proportion of phosphoric acid and of fluorine, an index to the degree of fossilization.

(*To be continued.*)